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Survey of Experimental Data of Thermophysical Properties for Difluoromethane (HFC-32)[1]

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A survey of experimental data for HFC-32 was prepared at the Institute of Thermomechanics in connection with planned experiments. In tabular form, surveys of thermodynamic, transport, and other property measurements, including *pvT* behavior, second virial coefficient, vapor pressure, saturation densities, critical parameters, heat capacities, speed of sound, thermal conductivity, viscosity, surface tension, refractive index, dielectric constant, and dipole moment, are presented. Tables include author(s) name(s), reference, year of publication, ranges of measurements, number of points, stated uncertainty, sample purity, and experimental method.

KEY WORDS: critical parameters; dielectric constant; difluoromethane; dipole moment; heat capacities; HFC-32; *pvT* behavior; refractive index; saturation densities; second virial coefficient; speed of sound; surface tension; survey; thermal conductivity; vapor pressure; viscosity.

1. INTRODUCTION

In connection with planned experiments, we have prepared a critical bibliography of experimental data and property equations for 13 thermophysical properties of difluoromethane, which is based on 134 literature sources published to the end of 1997. HFC-32 is an alternative refrigerant with application at low temperatures. Its mixtures with other refrigerants such as HFC-134a, HFC-125, HFC-152a, and HFC-143a are considered to be the most promising alternatives for HCFC-22 and the azeotropic mixture R-502.

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Table I. Survey of pvT Measurements of HFC 32

Author(s)	Year	Ref.	Pressure range (MPa)	δp $\pm$(kPa)	Temperature range (K)	δT $\pm$(mK)	Density range (kg · m^{-3})	$\delta \rho$ $\pm$(%)	Sample purity (mass%)	No. of Points	Method[a]
Malbrunot et al.	1968	2	0.8–20	1	248–473	30	20–1133	–	99.95	150	1, 2
Nishimura et al.	1992	3	0.15–6.6	0.8	290–370	10			99.98	95	3
Sato et al.	1992/1994	4, 5	3.3–9.8	2	322–420	7	111–850	0.1	99.998	69	4
Widiatmo et al.	1992	26	0.09–3.7	10	220–330	15	795–1215	0.2	99.998	14	
Qian et al.	1993	7	0.15–6.6	0.8	290–370	10	2.6–303	0.2	99.98	95	3
Defibaugh et al.	1993	8	0.24–9.8	0.5	242–372	10	4–1157	–	99.99	386	3, 5
Holste et al.	1993	9	1.5–72	0.02	150–375	10	451–1446	0.1	99.99	126	7
Baroncini et al.	1993	10	0.7–2.7	0.5	273–360	10	19.7–56.9	0.2 kg · m^{-3}	99.6	93	2
Grebenkov et al.	1994/1996	11		0.15%	280–350	20		0.3	99.99		1
Magee (Howley)	1994/1996	12	3.8–35	0.01–0.05%	142–396	30	708–1420	0.05	99.94	136	2
Bouchot and Richon	1994	13	0.12–9.5	3	253–333	20	2.7–1070	<0.2 kg · m^{-3}	>99.3	36	5
Fu et al.	1995	14	0.07–5.7	0.5	243–373	10	1.8–240		99.95	123	3
Fukushima et al.	1995	15	1.8–10	3	313–340	10	46.5–802	0.2	99.98	158	2
Takahashi et al.	1995	16	0.1–10	0.5	298–423	10	1.5–546	0.12	99.972	114	6
Lüddecke and Magge	1996	17	5.2–31.7	1	153–341	10	708–1417	0.1	99.94	74	2
de Vries	1997	18	0.02–20.6	0.1–0.2%	223–433	5–10		0.3–0.5	99.99	1194	3–5

[a] Methods: (1) variable volume method (Hg as confining medium); (2) constant volume method; (3) Burnett expansion method; (4) isochoric method with expansion; (5) vibrating tube densimeter; (6) oscillating disk viscometer adopted for density measurements; (7) continuously weighed pycnometer.

2. DATA SURVEY

Summaries of experimental measurements are presented in tabular form. (See Tables I to XIII.) In this section is presented additional information not included in survey tables. Authors usually also give correlation equations of the studied property comprising their own data only or a set of selected measurements and comparisons. More information on correlation expressions, comparisons, and available equations of state are given in Ref. 1.

Measurement techniques for vapor pressure comprised a wide range of methods depending on the pressure range. Most methods for the measurements in the low-pressure region, approximately below 200 kPa, are time-consuming and relatively inaccurate. Recently, three estimation procedures appeared in the literature which make it possible to extrapolate vapor pressure from the normal boiling temperature down to the triple point. They are applied by Lüddecke and Magee [17] using heat capacity measurements, by Tillner-Roth [33] employing a nonlinear regression analysis based on the Clausius–Clapeyron equation and a simple relation of the enthalpy of vaporization, and, finally, by Duatre-Garza and Magee [34] based on the measured internal energy changes.

The triple-point temperature and enthalpy of fusion were measured by Lüddecke and Magee [17] on three samples: $T_{tp} = 136.34 \pm 0.03$ K and

Table II. Second Virial Coefficients of HFC-32

Author(s)	Ref.	Year	Temperature range (K)	Uncertainty (%)	Method[a]
Dymond and Smith	19	1980	289–349	<10	1
			273–323	<10	1
Weber and Goodwin	21	1993	200–250	2-5	2
Qian et al.	7	1993	290–370	—	3
Bignell and Dunlop	78	1993	290–310	$\pm 2\ \mathrm{cm^3 \cdot mol^{-1}}$	
Sato et al.	5	1994	340–420	1	4
Hozumi et al.	22	1994	273–340	(1)	5
Defibaugh et al.	8	1994	373.14		3
			(268–373)		2
Zhang et al.	23	1995	290–370	<4	6
Sun et al.	48	1997	273–333	—	2
Yokozeki et al.	24	1997	180–550	0.1	7

[a] Methods: (1) from compressibility data by Fogg et al. [20] and Malbrunot et al. [2]; (2) estimated/calculated; (3) from graphical analysis of isotherms $(z - 1)\rho$ vs. ρ; (4) by fitting truncated virial equation of state; (5) from low pressure speed-of-sound measurements; (6) improved analysis of Burnett measurement; (7) analysis applying Stockmayer potential.

Table III. Vapor Pressure Measurements of HFC-32

Author(s)	Year	Ref.	Temperature range (K)	Pressure range (MPa)	Sample purity (mass%)	No. of points	Uncertainty δp (kPa)	Uncertainty δT (mK)	Methods[a]
Malbrunot et al.	1968	2	190.15–351.56	0.016–5.814	>99.95 mol	30	±0.1%	50	A, B
Kanungo et al.	1987	25	149.36–244.82	0.02–0.085	98.3	138[b]	0.01%	1	K
Nishimura et al.	1991	3	280–350		99.98	9	±0.85	10	E
Qian et al.	1992/1993	7	280–350	1–5.63	99.98	9	0.8	10	E
Widiatmo et al.	1992/1994	26, 27	219.97–324.98	0.09–3.27	99.998	25	2	15	F
Holcomb et al.	1993	28	295.28–348.63	1.57–5.46	99.9	25	±3.5	100	I
Weber and Goodwin	1993	21	208.36–237.38	0.05–0.21	99.98	27	0.03%	3–4	C
Zhu et al.	1993	29	273.39–347.29	0.82–5.32	99.95	32	0.5	10	E
Nagel and Bier	1993	30, 31	204.44–351.23	0.04–5.78	99.9	27	0.6	30	H
Baroncini et al.	1993	10	237.95–350.92	0.22–5.75	99.57	56	±0.5	10	H
Weber and Silva	1994	32	235.87–266.35	0.20–0.65	99.99 mol	17	0.0065	5	D
Defibaugh et al.	1994	8	268.15–348.08	0.69–5.41	99.99	18	0.04%	10	E
Sato et al.	1994	5	320.00–351.24	2.92–5.78	99.998	21	2	7	G
Bouchot and Richon	1994	13	253.07–333.39	0.40–3.95	>99.3	8	±5	20	J
Magee (Howley)	1994/1996	12	270–330	0.73–3.67	99.94 mol	7	±0.1%	30	H
Fukushima et al.	1995	15	277.59–350.95	0.93–5.74	99.98	57	±3	±10	H
de Vries	1997	18	223–350.15	0.11–5.65	99.99	139	0.5	±10	G

[a] Methods: (A) manometric; (B) static; (C) glass comparative ebulliometer; (D) metal comparative ebulliometer; (E) Burnett apparatus; (F) sinker method with magnetic suspension with a metallic bellows variable-volume cell; (G) isochoric method with expansion; (H) constant volume method; (I) vapor-liquid equilibrium apparatus; (J) vibrating tube densimeter; (K) differential manometry method.

[b] Values not presented.

Table IV. Saturation Densities of HFC-32

Author(s)	Year	Ref.	Temperature range (K)	$\delta\rho$ $\pm$(%)	Sample purity (mass%)	State[a]	No. of points	Method[b]
Malbrunot et al.	1968	2	249–352	0.2	99.95 mol	ρ'	16	1, 2
Widiatmo et al.	1992	26	275–330	0.2	99.998	ρ'	13	3
Holcomb et al.	1993	28	295–349	$\pm 0.5\ \mathrm{kg \cdot m^{-3}}$	99.9	$\rho' + \rho''$	25 + 25	10
Widiatmo et al.	1994	27	210–330	0.2	99.998	ρ'	25	3
Defibaugh et al.	1994	8	243–338	0.2	99.99	ρ'	21	4
			219–343			ρ''	28	
Sato et al.	1994	5	321–344	0.5	99.998	ρ'	2	5
			327–343			ρ''	4	
Higashi	1992/1994	36	336–351		99.998	$\rho' + \rho''$	17 + 17	6
Bouchot and *Richon*	1994	13	253–333	$\pm 0.3\ \mathrm{kg \cdot m^{-3}}$	>99.3	$\rho' + \rho''$	5 + 5	9
Kuwabara et al.	1992/1995	37	330–351	0.1–0.4	99.998	ρ'	17	6
			346–351			ρ''	13	6
Fukushima et al.	1992/1995	15	233–329	0.4	99.98	$\rho' + \rho''$	(11 + 8), 5	5, 8
			340–351	$\pm 3\ \mathrm{kg \cdot m^{-3}}$	99.98	ρ''	15	6
Magee	1994/1996	12	139–305	0.1	99.94	ρ'	13	5

[a] ρ', saturated liquid; ρ'', saturated vapor.

[b] Methods: (1) graphical extrapolation of isotherms to the vapor pressure curve; (2) pycnometric; (3) sinker with magnetic suspension; (4) isotherms extrapolated to the saturation boundary using the Tait equation; (5) intersection of isochores with vapor pressure curve; (6) meniscus disappearance; (7) *meniscus disappearance* and critical opalescence; (8) pyrex glass floats; (9) vibrating tube densimeter; (10) vapor–liquid equilibrium recirculation apparatus.

Table V. Critical Parameters of HFC-32[a]

Author(s)	Ref.	Year	T_c (K)	P_c (kPa)	ρ_c (kg · m^{-3})	Sample purity (mass%)	Method[b]
Malbrunot et al.	2	1968	351.52 ± 0.2	5830	429.6	99.95	VOMD
McLinden	38	1990	(351.54)	(5830)	(430)	—	—
Singh et al.	39	1991	(351.56)	(5814)	(429.61)	—	—
Kuwabara	37	1992/1995	351.255 ± 0.010	—	424 ± 1	99.998 mass	MD + CO
Nishimura et al.	3, 7	1992/1993	(351.255 ± 0.010)	5780 ± 2	—	99.98	CPV
McLinden et al.	40	1993	(351.36)	(5791)	(427)	—	—
Fukushima et al.	15	1992/1995	351.26 ± 0.03	5778 ± 3	425 ± 5	99.998 mass	MD + CO
Holcomb et al.	28	1993	(351.54)	(5830)	428.50 ± 1.83	99.9	CLVM
Nagel and Bier	30	1993	351.23 ± 0.06	5783 ± 6	420 ± 8	99.9	CO
Defibaugh et al.	8	1994	(351.36)	5792.7 ± 2.4	—	99.99	CVP
Schmidt and Moldover	41	1994	351.35 ± 0.02	—	419 ± 7	99.9 mol	MDR
Sato et al.	4, 5	1992/1994	(351.255 ± 0.010)	5784 ± 2.5	—	99.998 mass	CVP
Higashi	36	1992/1994	351.26 ± 0.02	5785 ± 9	427.5	99.98	MD + CO
Outcalt and McLinden	35	1995	(351.35)	(5795)	(427)	—	—
Yata et al.	42	1996	351.14 ± 0.10	—	—	>99.5	em VL
de Vries	18	1997	(351.26)	5781 ± 0.001	—	99	CVP
van Poolen and Holcomb	43	1997	351.33 ± 0.28	—	429.17 ± 1.30	—	IM 3D

[a] Values in parentheses follow from evaulation or are taken from other authors.

[b] Methods: MD, meniscus disappearance; CVP, p_c determined from the vapor pressure correlation; VOMD, visual observation of meniscus disappearance; CO, critical opalescence; MDR, meniscus disappearance and reappearance; CLVM, critical volume fraction method by Van Poolen; em VL, empirical method using experimental data on vapor–liquid equilibrium; IM 3D, intersection method assuming 3D Ising-like system.

Table VI. Speed of Sound Measurements of HFC-32

Author(s)	Ref.	Year	Phase	Temperature range (K)	Pressure range (MPa)	No. of points	Uncertainty in w (%)	Sample purity (mass%%)	Method[a]
Takagi	44	1993	Liq	243-373	p_s-35	120	0.2	99.96	1
Hozumi et al.	22	1994	Gas	273-343	0.02-0.250	67	0.01	99.99	2
Grebenkov et al.	45	1994	Liq	286-341	p_s-10.3	30	0.2	—	3
Beljaeva et al.	46	1995			same date as in Ref. 45				
Hozumi et al.	47	1996	Gas	308-343	to 0.500	44	0.006	99.99	2
Sun et al.	48	1997	Gas	273-333	0.048-0.390	38	0.1	—	4

[a] Methods: (1) echo technique operated at 2 MHz with a fixed path interferometer; (2) spherical acoustic interferometer; (3) impulse method at frequency of 2.1 MHz; (4) variable-path acoustic interferometer.

Table VII. Heat Capacities of HFC-32

Author(s)	Year	Ref.	Type[a]	Temperature range (K)	Pressure range (MPa)	No. of points	Sample purity (mass%)	Uncertainty (%)	Method[b]
Yomo et al.	1992/1994	49	c_p	275–315	2.1–3.0	41	99.936	±0.35–0.6	1
Weber and Goodwin	1993	21	c'_p, c''_p	200–250	p_s	6 + 6	—		2
Defibaugh et al.	1994	8	c'_p, c''_p c'_v, c''_v	200–T_c	p_s	a27	—		2
Lüddecke and Magee	1996	17	c'_v	145–342	p_s	95	99.94	±0.7	3
			c_v	152–341	5.8–31.7	73	99.94		3
Beljajeva et al.	1995	46	c'_p	250–325	p_s	16	—		2
			c_p	285–325	1.5–3.5	29			2
				Ideal-gas heat capacity					
Rogers	1974	50		0–1500 K	Calculated from spectroscopic data				
JANAF tables	1985	51		0–6000 K	Calculated from spectroscopic data				
McLinden	1990/1993	38, 40		150–600 K	Correlated data from Ref. 50				
Defibaugh et al.	1994	8		200–400 K	Calculated from spectroscopic data				
Hozumi et al.	1994/1996	22, 47		200–400 K	Calculated from low pressure speed of sound mesurements				
Yokozeki et al.	1999	24		120–1000 K	Complete analysis of spectroscopic date				
Sun et al.	1997	48		273–333 K	Calculated from low pressure speed-of-sound mesurements				

[a] ′, saturated liquid; ″, saturated vapor.

[b] Methods: (1) flow calorimeter; (2) calculated; (3) adiabatic calorimeter.

Table VIII. Thermal Conductivity Measurements of HFC-32

Author(s)	Year	Ref.	Phase[a]	Temperature range (K)	Pressure range (MPa)	No. of points	Uncertainty (%)	Sample purity (mass%)	Method[b]
Tauscher	1969	52	Liq	148–298	0.01–2			--	THWM
Papadakai and Wakeham	1993	53	Sat	205–303	p_s	10	±1	99.9	THWM (TaCW)
			Liq						
Geller and Paulaitis	1994	54	Gas	255–383	0.1–5	42	⩽1		
			Sat	245–344	p_s	12	Near CP		MSSHWM
			Scr	352–428	4.1–19.7	67	5		
Geller et al.	1994	55	Gas	253–373	0.1–5.3		⩽1	--	MSSHWM
Grebenkov et al.	1994/1996	11	Sat	275–403	1.67–12.9	96	To 3.5	99.99	COAC
Tanaka et al.	1995	56	Gas	283–333	0.1–3.93	53	±1	99.99 mol	THWM (2PtW)
Ro et al.	1995	57	Liq	223–323	2–20	24	±2	99.9	THWM (bPtW)
Assael and Karagiannidis	1995	58	Liq	253–313	0.6–17.6	27	±0.5	99.98	THWM (anTaW)
			Sat + Liq			4			
Gross and Song	1996	59	Liq + Gas	233–345	0.1–6.18	80	1.6–2	99.8	THWM (pV)
Yata et al.	1996	60	Liq	253–324	2–30	27	1	>99.5	THWM
Sun et al.	1997	61	Gas	255–342	Near p_s	20	3	99.95	THWM (2-anTaW)
Gao et al.	1998	62	Sol + Liq	323	p_s	13	2	—	THWM (bPtW)

[a] gas, gaseous; Liq, Liquid; scr, supercritical region; sol, solid; sat, saturated state.

[b] Methods: THWM, transient hot-wire method; anTaW, anodized tantalum wire; TaCW, coated tantalum wire; 2PtW, two Pt wires; bPtW, single bare Pt wire; pV, with polarization voltage; MSSHWM, modified steady-state hot-wire method; COAC, steady state method of coaxial cylinders.

Table IX. Viscosity Measurements of HFC-32

Author(s)	Year	Ref.	Phase[a]	Temperature range (K)	Pressure range (MPa)	No. of points	Uncertainty (%)	Sample purity (mass%)	Method[b]
Phillips and Murphy	1970	63	η'	200–287	0.01–2	10	—	99.9 mol	CTV
Oliveira and Wakeham	1993	64	η'	232–343	0.21–5.1	19	±0.6	—	VWV
			η''	252–333	0.37–3.96	14	±0.7		VWV
Ripple and Matra	1993	65	η'	251–293	0.36–1.48	10	3–5	—	CTV
Assael et al.	1994	66	Liq	273–313	p_s–15.5	26	±0.5	99.98	VWV
			Sat	273–313	p_s	5			Calculated
Dunlop	1994	67		298.15		1	0.3	—	CTV
Grebenkov et al.	1994	11	Liq	290–316	To 16	54		99.99	FCV
Takahashi et al.	1995	16	Gas	298–423	0.10–5.4	58	±0.3	>99.972	ODV
Geller et al.	1995	68	Gas	253–363	0.45–5.4	51	<±1.2	—	MCTV
			η', η''	253–348	p_s	10+10			Calculated
Sun et al.	1996	69	η''	233–333	p_s	21	<3	—	CTV
Heide	1996	70	η'	223–333	$p_s+0.1$	12	2	99.4	FBV

[a] ′, saturated liquid; ″, saturated vapor.

[b] Methods: VWV, vibrating-wire viscometer; FBV, falling-ball viscometer; ODV, oscillating-disk viscometer; MCTV, modified capillary-tube viscometer; CTV, capillary-tube viscometer; FCV, falling-cylinder viscometer.

Table X. Surface Tension Measurements of HFC-32

Author(s)	Year	Ref.	Temperature range (K)	No. of points	Uncertainty ($mN \cdot m^{-1}$)	Sample purity (mass%)	Method[a]
Okada and Higashi	1994	71	273-333	13	±0.2	99.98	DCRM
Schmidt and Moldover	1994	41	297-350	10	(±0.15)	99.9	DCRM
Zhu and Lu	1994	72	268-334	13	—	99.95	DCRM

[a] DCRM, differential capillary-rise method.

Table XI. Refractive Index of HFC-32

Author(s)	Year	Ref.	Temperature range (K)	State	No. of points	Wavelength (nm)	Sample purity (mass%)	n_c
Schmidt and Moldover	1994	41	296-T_c	Sat. liq	14	633	99.9	1.0828
				Sat. vap	14			
Yata et al.	1996	42	313-T_c	Sat. liq	28	545	99.5	1.0854
				Sat. vap	28			

Table XII. Dielectric Constant of HFC-32

Author(s)	Year	Ref.	Phase	Temperature range (K)	Pressure range (MPa)	No. of points	Uncertainty (%)
Tremaine and Robinson	1973	73	Sat. liq	152-224	p_s	8	±0.8
Meyer and Morrison	1991	74	Gas	308-411	0-0.45	54	n.a.
Nieto de Castro et al.	1995	75	Liq	208-303	2-17	165	0.1

[a] All measured with capacitance cells.

Table XIII. Dipole Moment of HFC-32

Author(s)	Year	Ref.	Method	μ (D)
Lide	1951	76	From Stark effect measurements	1.96 ± 0.02
Kawaguchi and Tanaka	1977	77	From analysis of Stark spectrum	1.9785 ± 0.0021
Meyer and Morrison	1991	74	Calculated from dielectric measurements	1.978 ± 0.007

$\Delta H_{\text{vap}} = 4356 \pm 130$ kJ · mol^{-1}. The triple-point pressure was estimated by Tillner-Roth to be 50.70 ± 0.14 Pa [33], while that calculated from the equation of state by Outcalt and McLinden [35] is 46.9 Pa. This value is in good agreement with the value of 46.5 Pa recently evaluated by Duarte-Garza and Magee [34] using the internal energy method.

A small difference exists in the critical parameters used in equations of state derived by Japanese and U.S. researchers:

	T_c (K)	p_c (MPa)	ρ_c (kg · m^{-3})
Japan	351.255	5.780	424
U.S.A.	351.35	5.795	427

The low-pressure speed-of-sound data were used to derive the second acoustic virial coefficients, ideal-gas heat capacities, and the second virial coefficients. Discrepancies among various sets of thermal conductivity indicate that the quoted uncertainties are overestimated. There is a suspicion that not all measurements of thermal conductivity were published until recently since the mutual discrepancies exceeded the stated accuracy. Large discrepancies observed in the measured liquid viscosities are attributed mostly to the impurities in the samples, the electrolyte effect, and the incapability of instruments to be calibrated with water.

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